IV.D.1 SOFC Modeling at PNNL

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Objectives

- Develop and validate multi-physics modeling tools to simulate solid oxide fuel cell (SOFC) stack performance.
- Utilize computational techniques for the mitigation of performance degradation and optimization of modular SOFC stack and system designs.
- Obtain necessary material properties to support the development and optimization of SOFC designs through modeling.
- Disseminate/transfer modeling tools to Solid State Energy Conversion Alliance (SECA) industry teams and Core Technology Program (CTP) members.

Accomplishments

- Developed a finite element-based code, called SOFC-Multi Physics (SOFC-MP), to solve the coupled flow, electrochemistry, and heat transfer solution in fuel cell stacks under steady-state conditions.
- Co-developed with MSC Software a stack design tool, called Mentat-Fuel Cell (Mentat-FC), for SOFC model generation, integration/solution with SOFC-MP, and structural analysis using the MARC finite element mesh and modeling capabilities.
- Provided training for SOFC modeling tools and techniques through a demonstration seminar at the April 2005 SECA workshop, comprehensive technical programs hosted at PNNL covering the full suite of SOFC modeling tools, individual student training and collaboration at PNNL, and individual telephone/email support.
- Distributed the SOFC-MP and Mentat-FC software packages to multiple industry teams and CTP university researchers for modeling and development of SOFC stacks.
- Established a methodology to assess glass-ceramic seal failure. A continuum damage mechanics model based on the experimental stress/strain response was developed for G18 sealing glass. The damage model was implemented in MSC MARC and used for SOFC stack stress analysis to predict accumulated damage and failure of the seals under thermal-mechanical loading. The methodology was extended to predict seal damage accumulation in stacks due to thermal cycling processes.
- Developed a probabilistic-based component design methodology for the SOFC stack. This method takes into account the randomness in SOFC material properties as well as stresses arising from different manufacturing and operating conditions.
- Developed an integrated modeling/experimental framework to predict the life of SOFC interconnect materials. Oxide scale properties were evaluated experimentally and the effects of interconnect oxide growth on interfacial structural integrity during isothermal cooling was studied.
- Initiated a design basis document in collaboration with the American Society of Mechanical Engineers (ASME) and Oak Ridge National Laboratory (ORNL) to provide industry teams with technical guidance on materials characterization, constitutive models, modeling techniques, failure analyses, and software usage to support SOFC design and development efforts.
- Developed modeling methodologies and constitutive models based on experimental characterizations to evaluate the time-dependent mechanical response of stack components. The models can quantify the effect of creep in metallic components and glass-ceramic seals on stack deformations and cell component stresses during operation and shutdown. A homogenization model to predict glass-ceramic seal properties as a function of composition was developed and implemented.
- Established a methodology to assess interconnect scale growth and effect of the associated electrical resistance increase on stack performance. The capability enables evaluation of the long term behavior of prospective interconnect materials with respect to thermal and electrical stack performance.
- Supported development of a standardized SOFC cell geometry for use in the SECA program to evaluate materials and technologies within a common testing platform.

Introduction

In order to efficiently develop and optimize planar SOFC stacks to meet technical performance targets, it is desirable to experiment numerically with the effects of geometry, material properties, operational parameters, and thermal-mechanical loading. The computations with representative baseline designs, validated by experimental data, have been used to develop better understanding of the stack behavior while avoiding costly and time-consuming experiments. In order to model the coupled physics associated with an SOFC stack, the simulation tool SOFC-MP was developed. This modeling tool combines the versatility of a commercial multi-physics code and a validated electrochemistry calculation routine to predict the gas flow distributions, current distribution, temperature field, and power output for stack-level simulations. The fundamental building blocks of the modeling and simulation tools are electrochemical models, heat and mass transfer simulations, computational mechanics, and experimental data.

The multi-physics modeling tools developed were then used in studying a wide range of design criteria as well as current material development and degradation challenges. A probabilistic-based component design methodology was developed, which takes into account the randomness in SOFC material properties as well as stresses arising from different manufacturing and operating conditions. For SOFC materials and stack development, the time-dependent mechanical response of seal and interconnect components were considered to predict the impact on stack performance and component stresses. Interconnect scale growth was evaluated for both its mechanical durability to resist growth-induced spallation as well as its influence on the cell electrochemistry. The modeling tools were also used to evaluate issues expected to be problematic for cell scale-up such as high stresses and loss of contact in the stack. The developed design methodology and stack analytical procedures are currently being incorporated into a design basis document for distribution within the SECA program.

Approach

The following technical approach has been taken in the modeling task to meet program goals:

- Maintain, enhance, and provide guidance for the integrated modeling tools developed under the SECA CTP for evaluating fuel cell stack design concepts by the industry teams.
- Investigate the effects of materials degradation on cell performance and life.
- Investigate the effects of cell geometric design, material property distributions, and operating conditions on SOFC reliability.
- Perform material experiments for property data essential to constitutive and numerical model development.

Results

Increased Usage of Modeling Tools

The modeling tools and techniques developed at PNNL played a greater role in continued support of SOFC technology development for the SECA industry and university team members:

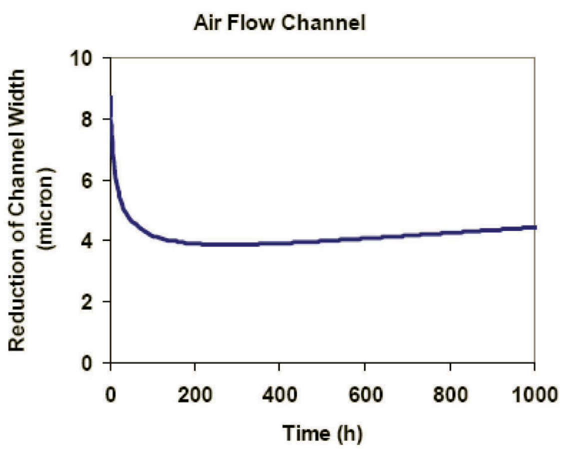
- The SOFC-MP and Mentat-FC modeling tools were delivered to GE Energy, Delphi, and West Virginia University with accompanying technical support for usage and operation.
- The developed modeling tools were used for collaboration with the University of Cincinnati to study the performance of their glass sealant in a realistic SOFC cell. Other university participants from West Virginia University, Carnegie Mellon University, and Georgia Tech will participate in summer internships to learn about SOFC modeling.
- The modeling tools were used for design of the SECA test cell. The pressure drop analysis showed that a serpentine geometry had small air pressure drop <10" H_2O for 4-cm tests cells, but was excessive if used for realistic cells sizes. In contrast, a non-serpentine channel design could be used for up to a 30-cm cell with comparable pressure drops. The thermal analysis for the serpentine and cross-flow geometries showed that the entire structure was nearly isothermal, and the structural analysis is now in progress.
- PNNL has co-led the development of the design basis document which will serve as a modeling and analysis guide for SECA members. Contributions regarding document objectives, modeling approaches, modeling tool usage, and failure analyses have been incorporated, and detailed modeling descriptions continue to be added to the analysis sections.

Modeling of Time-Dependent Material Properties

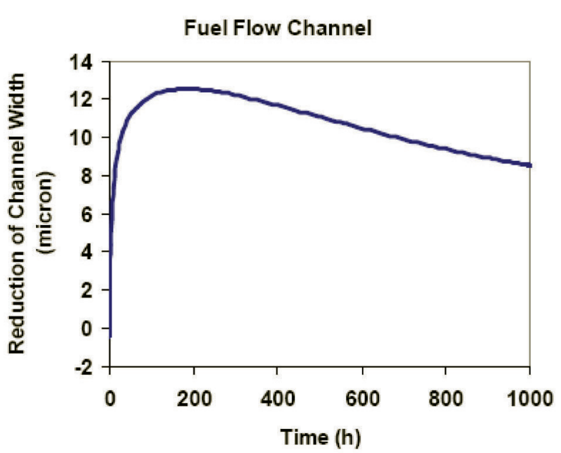
The use of time-dependent creep and stress-relaxation behaviors of cell materials was investigated and found to have a great influence on the stress predictions for the components.

- For the ferritic interconnect, modeling of the stack deformation during long periods of operation were simulated. The coefficient of thermal expansion (CTE) mismatches were found to cause cell bowing which changed with time and contributed to changes in the gas channel heights (Figure 1); however, significant beneficial reduction in positive electrode/electrolyte/negative electrode (PEN) and seal stresses were also observed.

- The effect of oxide scale growth over time on the scale strength and stress state was quantified using indentation tests and simulations.
- For the G18 sealant, experimental creep tests were performed to determine the secondary creep strain rate dependency on stress and temperature (Figure 2). The data was implemented into the previously developed continuum damage mechanics model to predict the material response at different strain rates, and the effects of creep/relaxation on cell stresses were evaluated for different operating and shutdown durations.
- The glass-ceramic constitutive model was also extended to include material property predictions for the homogenized sealant behavior based on the volume fractions and properties of the ceramic and glassy phases. The model can be used to evaluate the effect of different seal material properties on stack stresses during operation and shutdown.



(a)



(b)

FIGURE 1. Effect of Interconnect Creep on Flow Channel Heights for 1,000 Hours Operating Duration

Modeling for Issues Related to Scale-Up

The desired use of SOFCs in megawatt-scale power applications using coal-based fuels provided motivation to study scale-up of cell dimensions. Larger cells are expected to have greater issues with thermal management for mechanical reliability, electrochemical variability across the cell, and maintenance of the electrical contact path. The modeling tool capabilities were extended to evaluate these issues:

- Cell sizes of 10-20-cm were evaluated for co-, counter- and cross-flow configurations, where the average stack temperature was kept constant by varying the inflow gas temperatures. The results (Table 1) indicated that the temperature difference increased by 70-100 K and peak stresses in the anode were significantly increased.

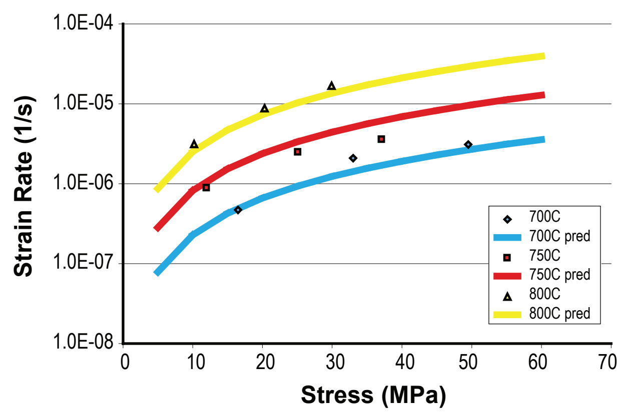


FIGURE 2. Secondary Creep Strain Rate for 4-hour Heat Treated G18 Glass-Ceramic as a Function of Stress and Temperature

TABLE 1. Predicted Temperatures, Peak Anode Principal Stress, and Peak Interconnect von Mises Stress for Cells of Different Sizes and Flow Configurations

Variables	Co-flow		Counter-flow		Cross-flow	
	10x10	20x20	10x10	20x20	10x10	20x20
Max T (°K)	1049	1120	1054	1155	1051	1170
Min T (°K)	970	898.5	967.1	847.9	970.3	857.3
σ_{IC} (MPa)	20.95	36.03	20.83	63.21	20.23	50.39
σ_{anode} (MPa)	10.30	36.06	13.67	63.87	9.612	51.0

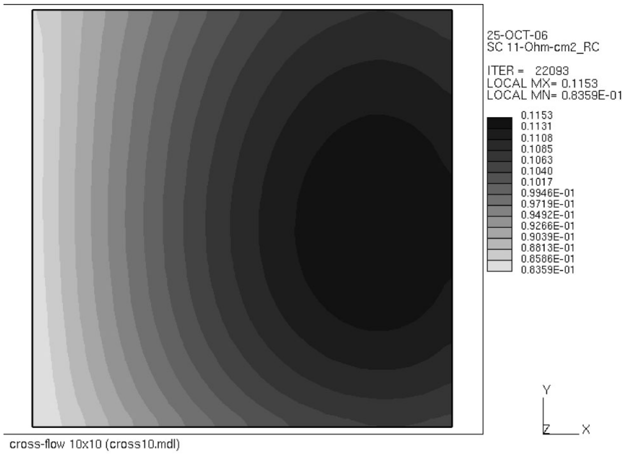


FIGURE 3. Predicted Cathode-Side Interconnect Electrical Resistance Distribution (Ω-cm^2) for a Stack Operated at 750°C, 0.6 A/cm^2 for 10,000 Hours

- The greater range of operating temperatures in a large cell are expected to affect the rate of thermally driven mechanisms such as scale growth. The growth of the interconnect scale and corresponding increase in the electrical resistance was simulated, and it was found that the local area specific resistance (ASR) varied from 0.83-0.12 Ω-cm^2 across the active area (Figure 3) with an increase of 26°C in the cell temperature difference after 10,000 hours of operation.
- Each cell in the stack must maintain electrical contact with the interconnect by a combination of mechanical bonding and compressive loading. Loss of contact through deformations or debonding and the corresponding impact on performance due to greater ohmic losses was simulated by modeling the stack electric field. The modeling capability was used to quantify a cell's total ohmic loss due to different corrugated interconnect geometries and loss of cathode contact due to mechanical bowing of the PEN.

Conclusions and Future Directions

In the last year, the modeling tools had greater usage and were enhanced with additional capabilities to address durability issues. Future modeling activities will continue to focus on reliability, degradation, time-dependent response, and scale-up issues:

- Continue to improve the modeling tools to meet the needs of the SECA program. Continue to increase the usage of the tools by the industry and academic teams.
- Continue to add improved material models and numerical procedures to the modeling tools for simulation of time-dependent response and reliability.
- Continue modeling to improve bond strengths of the oxide and protective coating layers for ferritic stainless steel interconnects.
- Evaluate thermal management needs, influence of high pressure electrochemistry, and reliability of seal/cell structures during cell scale-up.
- Continue to support development of a robust test cell design.
- Evaluate the mechanical requirements for successful fabrication using refractory glass sealants and low-temperature sintering of cathode contact materials for reliable interconnection during operation and shutdown.
- Continue to develop seal property predictions via homogenization methods to identify reliable composite seal structures and compositions for stacks.
- Develop analytical methods to evaluate the time-dependent mechanical behavior (creep, thermal fatigue, loss of interconnect contact) of fuel cell stacks/components and corresponding influence on electrochemical performance.

FY 2007 Publications/Presentations

1. MA Khaleel, KP Recknagle, X Sun, BJ Koeppel, EV Stephens, BN Nguyen, KI Johnson, VN Korolev, JS Vetrano, and P Singh, "Recent Development of Modeling Activities at PNNL," presented at the SECA Core Technology Program Peer Review, Philadelphia, PA, September 12–14, 2006.

2. KP Recknagle, BJ Koeppel, X Sun, JS Vetrano, ST Yokuda, DL King, P Singh, and MA Khaleel, "Analysis of Percent On-Cell Reformation of Methane in SOFC Stacks and the Effects on Thermal, Electrical, and Mechanical Performance," presented at the Fuel Cell Seminar 2006, Honolulu, HI, November 13–17, 2006.

3. X Sun, W Liu, J Vetrano, G Yang, MA Khaleel and M Cherkaoui, "Life Prediction of Ferritic Stainless Steel Interconnect under Thermal Stress and Oxide Growth Stress," presented at the Fuel Cell Seminar 2006, Honolulu, HI, November 13–17, 2006.

4. W Liu, X Sun, and MA Khaleel, "Fracture Failure Criteria of SOFC PEN Structure," presented at the 31st International Conference on Advanced Ceramics and Composites, Daytona Beach, FL, January 21–26, 2007.

5. BN Nguyen, BJ Koeppel, and MA Khaleel, "Design of a Glass-Ceramic Seal for Solid Oxide Fuel Cell Applications by Means of a Homogenization Approach," presented at the ASME Applied Mechanics and Materials Conference, Austin, TX, June 3–7, 2007.

6. X Sun, W Liu, and MA Khaleel, "Effects of Interconnect Creep on Long-Term Performance of a One-Cell Stack," PNNL–16342, Pacific Northwest National Laboratory, Richland, WA, 2007.